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A double-layer heating method to generate high temperature in a two-stage multi-anvil apparatus[*]

Bo Peng(彭博), Zili Kou(寇自力)[†], Mengxi Zhao(赵梦溪), Mingli Jiang(姜明莉),
Jiawei Zhang(张佳威), Yipeng Wang(王义鹏), and Lu Zhang(张陆)

Institute of Atomic and Molecular Physics, Sichuan University, Chengdu 610065, *China*

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A new heating method is proposed to increase the cell temperature of the 6–8 type multi-anvil apparatus without reducing the volume of the sample chamber. The double-layer heater assembly (DHA) has two layers of heaters connected in parallel. The temperature of the cell was able to reach 2500 °C by using 0.025 mm rhenium foils, and the temperature limit was increased by 25% compared with that of the traditional single-layer assembly. The power–temperature relationships for these two assemblies with different sizes were calibrated by using W/Re thermocouple at 20 GPa. When the volume of samples was the same, the DHA not only attained higher temperature, but also kept the holding time longer, compared to the traditional assembly. The results of more than ten experiments showed that the new 10/4 DHA with a relatively large sample size (2 mm in diameter and 4 mm in height) can work stably with the center temperature of the sample cavity exceeding 2300 °C under the pressure of 20 GPa.

Keywords: double-layer heater, high pressure and high temperature, multi-anvil apparatus

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1. Introduction

Pressure and temperature are two essential thermodynamic parameters that determine the state of matter and affect the physical properties of matter. High pressure and high temperature (HPHT) technology is not only an effective way to find new materials,[1,2] but also is an important approach to synthesize superhard materials like man-made diamond and cubic boron nitride (cBN).[3–5] What is more, physical conditions inside the Earth can be simulated by HPHT technology, which helps to develop a more accurate picture about the interior of Earth.[6]

At room temperature, the achievable pressures of a multi-anvil apparatus are 30 GPa and 120 GPa respectively by using tungsten carbide (WC) and sintered diamond (SD) cubic anvils.[7,8] Under the condition of lower pressure (∼ 10 GPa), graphite is usually used as a heating material because of its stability and low cost. However, under high pressure (over 10 GPa) graphite transforms to diamond even at relatively low temperature, which eliminates the use of this material.[9] Accordingly, the metals with high melting points, such as tantalum (Ta) and rhenium (Re), are widely used as heating materials in high pressure experiments.[14] The maximum attainable temperatures are approximately 2000 K and 2700 K for Ta and Re, which are lower than their melting points (∼ 3000 ℃).[6,15] When refractory metals were used as heaters, Ito *et al.* proposed that a small amount of hydrogen, from residual water in the assembly, dissolved into the metal heaters, which lowered the melting temperature of the refractory metals greatly.[6] This effect may lead to the lower attainable temperature of a metal heater even if its melting point is high. Recently, boron-doped diamond was used as a heating material in a multi-anvil high-pressure apparatus and it generated extremely high temperature (∼ 3700 ℃).[10] However, the application of boron-doped diamond was limited by the small size of samples (0.7 mm in diameter). The maximum attainable temperatures with different heating materials are also affected by the type and size of the thermal insulator that is composed of lower thermal conductivity materials such as ZrO_2 and $LaCrO_3$.

Wang *et al.* designed and optimized a traditional single-layer heater assembly (SHA) based on DS6 × 8 MN 6–8 type multi-anvil cubic press and realized 2000 ℃ at 18 GPa.[16] However, a drawback of the SHA is that the size of sample is small (2 mm in height and 1.2 mm in diameter), and the small size limits the application of samples and the macrocosmic tests of the sample's mechanical properties.

In this paper, we increase the cell temperature from 2000 ℃ to 2500 ℃ by improving and optimizing the traditional assembly and simultaneously realize the relatively larger cell volume at 20 GPa. Compared with the single-layer heater assembly (SHA), this double-layer heater assembly (DHA), which is featuring in compatible longer holding time and the stable heating process, helps to improve the properties of some sintered samples and provides a reference for the improvement of the other assemblies with different sizes.

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[†] Corresponding author. E-mail: kouzili@scu.edu.cn

2. Assembly and experiments

2.1. The design of new assembly

The resistance of metals such as Re and Ta increases with temperature, making the heating more concentrated on the center. This situation leads to a rapid rise of the cavity temperature at the high-temperature stage, which is easy to cause the sudden fusion of the metal heating tubes and the failure of heating.

The total resistance of a parallel circuit is lower than its individual resistance, just like Fig. 1. So we added another heating tube to the normal assembly and connected two heaters in parallel to reduce the resistance of the metal heaters and improve the stability of the heating process. We chose the aluminum oxide (Al_2O_3) tube with 0.2 mm thickness to separate the two heaters and achieve a parallel connection. The schematic diagrams of two kinds of cell assemblies are shown in Fig. 2.

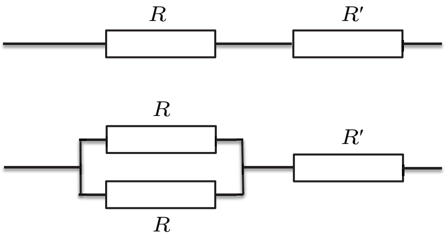

Fig. 1. The R and R' indicate the resistance of the heaters and the total resistance of the other part of the assemblies, respectively. $P = I^2(R+R')$, under the same heating power, the total resistance decreases and the total current augments, which increases the heat generated in R' and decreases the heat generated in the heaters. So the method of double-layer heaters can slow down the rapid rise of the cavity temperature.

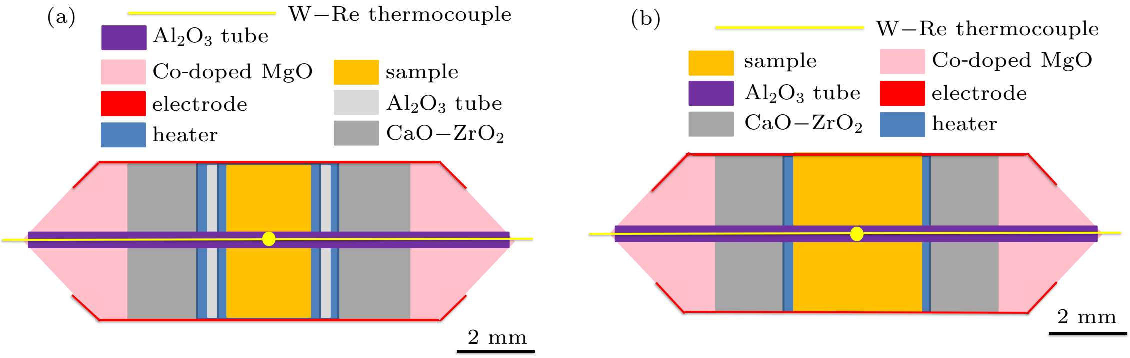

Fig. 2. The schematic diagrams of two assemblies. (a) The schematic diagram of the DHA. Two layers of heaters were insulated by an aluminum tube, and the two heaters were connected with two 0.2 mm Ta electrodes in the assembly. (b) The schematic diagram of the SHA. It is a normal 10/4 assembly which is used widely.

2.2. Heating experiments

Table 1 summarizes the runs of different assemblies and dissimilar sizes at 20 GPa. All temperature calibration experiments were conducted in a DS6 × 8 MN 6–8 type multi-anvil cubic press (Zhangjiakou Exploration Machinery Factory, China) located in Sichuan University.[11,12,19] The cubic tungsten carbide (WC) blocks with 4 mm truncation edge-length were used as the second-stage anvils to generate high pressure. The octahedral semi-sintered cobalt-doped magnesia (MgO) with length of 10 mm acted as the pressure medium.[13] The homemade calcium oxide–zirconia (CaO–ZrO_2) solid solution was used as the insulation material.[17] The Re foils were used as heaters and the Ta foils were used as electrodes in all runs. A CaO–ZrO_2 solid solution was used as the sample in all runs. The temperature at the center of the sample lower than 2300 °C was directly monitored by the W97Re3–W75Re25 thermocouple, while the temperature higher than 2300 °C was estimated based on a linear extrapolation of the power–temperature relationship.

Table 1. Summary of the heating experiments. The sample size is given in the order of diameter and height. The size of the thermal insulator tube is given in the order of thickness and height.

Assembly	Type No.	Sample size/mm	Thickness of heater/mm	Size of thermal insulator/mm	P/GPa	Highest temperature/°C
DHA	1	2.5/4	0.025	0.85/4	20	2000
DHA	2	2.3/4	0.025	0.95/4	20	2200
DHA	3	2/4	0.025	1.1/4	20	2500
DHA	4	2/3	0.025	1.1/3	20	2500
SHA	7	2.5/4	0.025	0.85/4	20	1800
SHA	8	2.3/4	0.025	0.95/4	20	1850
SHA	9	2/4	0.025	1.1/4	20	1950
SHA	10	2/3	0.025	1.1/3	20	2000
SHA	11	2/4	0.05	1.1/4	20	1950

3. Results and discussion

Figure 3 displays the power–temperature relationships of the runs with different sizes. Figures 3(a)–3(c) show the contrasts of type No. 1 and type No. 7, type No. 2 and type No. 8, type No. 3 and type No. 9, respectively. The results of those comparative experiments indicated that the DHA can attain higher temperature under the conditions of the same heater, the same thermal insulation material, and the same sample size. The DHA reduces the heating rate, which makes the heating process more stable. The heating rate of the DHA is lower than that of the SHA because of its smaller resistance of heaters in all runs. Figure 3(d) displays the power–temperature relationships of the heaters with different sizes which are for type No. 3 and type No. 11. The SHA with a 0.05 mm Re heater has a similar heating rate with the DHA with two 0.025 mm Re heaters. However, the DHA reaches higher temperature, which means that the simple use of thicker metal heaters in an assembly cannot generate higher temperatures. All relationships can be fitted by a polynomial function.

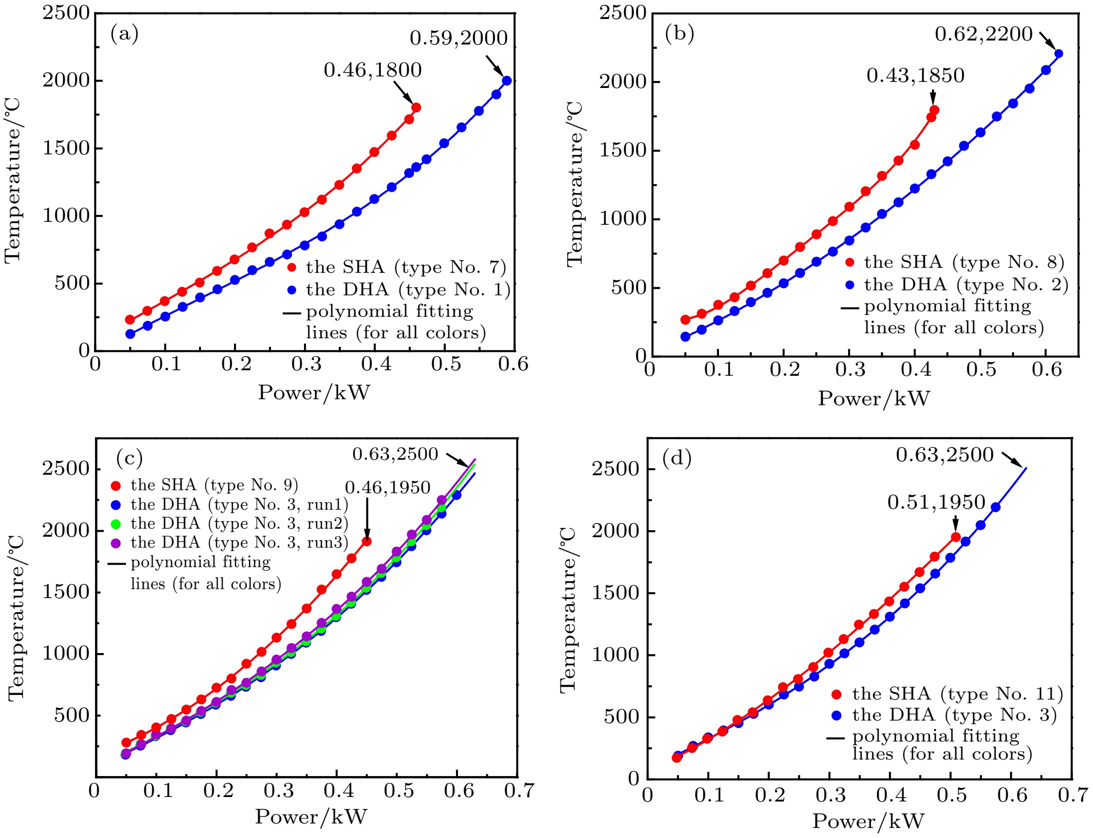

Fig. 3. The power–temperature diagrams of assemblies. (a)–(c) The relationships for the sample diameters of 2.5 mm, 2.3 mm, and 2 mm, respectively. (d) The relationships for type No. 3 and type No. 11.

We tested the holding time of the two best assemblies which are type No. 3 and type No. 9 many times. The results showed that the DHA can keep the high temperature longer than the SHA. Table 2 lists the holding time of experiments that carried out with the two assemblies.

Figure 4 shows the SEM images of the recovered assembly. A amount of dendrite crystal was found in the center area in Fig. 4(b), which indicated that the partial fusion took place in the sample.[20–22] The melting point of ZrO_2–CaO solid solution is approximately 2450 ℃,[18] which is consist with the highest temperature that obtained from the linear extrapolation of the power–temperature relationship of the DHA.

Table 2. The statistics of the temperatures and the average holding time of two assemblies.

Assembly	Temperature/℃	Holding time				
		Experiment times	Less than 3 min	3–5 min	Over 5 min	Average duration/min
SHA	2000	10	4	6	0	3.6
DHA	2000	10	0	6	4	5.3
	2300	10	3	7	0	3.2

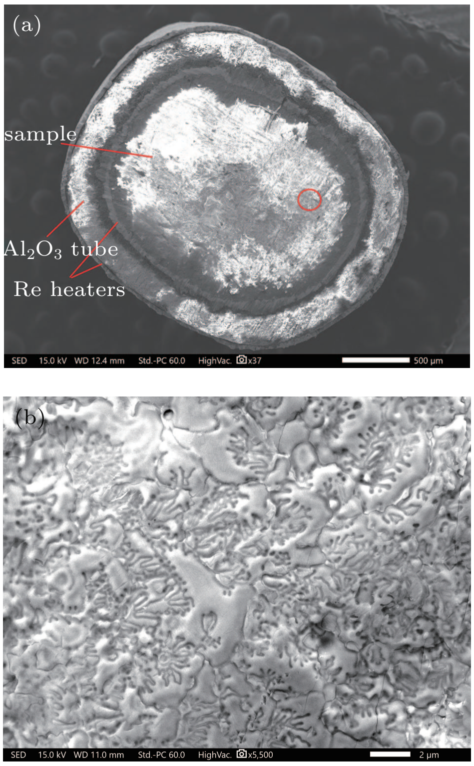

Fig. 4. The SEM images of recovered sample. (a) The image of recovered sample of the DHA (type No. 3). (b) Enlarged image of red circle in (a), the dendritic texture indicates the melting of the sample.

4. Conclusion

In this paper, we designed a double-layer heater assembly which can rise the temperature from 2000 ℃ to 2500 ℃ by using 0.025 mm Re heaters in a DS6 × 8 MN 6–8 type multi-anvil cubic press. Compared with the traditional single-layer assembly, the new assembly has a large cavity volume and a more stable heating process. Its thermal insulation performance was tested for many times, and the results showed that the DHA can keep longer holding time. The larger sample is conductive to the macrocosmic tests of its mechanical properties, and moreover, the longer holding time can contribute to improve the property of some materials synthesized by high pressure and high temperature technology.

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